

Amorphous silicon- from doping to multi-billion dollar applications.

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PACS code: 71.23 Cq, 73.61 Jc.

Keywords: amorphous silicon, doping, field effect, thin film transistor.

Abstract

In this paper, we recount the history leading up to the landmark paper by Spear and LeComber in 1975, which showed, contrary to thought at the time, that it was indeed possible to incorporate substitutionally pentavalent and trivalent impurities into a tetrahedral amorphous semiconductor. This work provided the basis for a multi-billion dollar business with products which are ubiquitous.

1. Introduction

Amorphous Silicon (a-Si) is the basis of a multi-billion dollar market in diverse applications such as active matrix liquid crystal displays (AMLCD), electro-photography, image sensors, solar cells, etc. The market for these products is expanding rapidly especially as the AMLCD displays are beginning to displace CRT's for TV with screen sizes of around 24" for less than US\$1,000.00. The origins of this remarkable commercialization success can be traced to the work at Standard Telephone Labs. (STL), Harlow, UK (1965-1969) and from 1970 at The University of Dundee (Scotland); at Dundee, the group headed by Professor Walter Spear, demonstrated not only the doping of amorphous silicon with phosphorous and boron in 1975 but the group also developed the

field effect technique (or thin film transistor-TFT) using a-Si/SiN_x combination in 1973, thus opening the way to the commercialization of this versatile semiconductor.

This paper is meant to commemorate the 30th anniversary of doping reported by Spear and LeComber. I am honored that the conference committee has asked me to recount the early history of a-Si.

2. a-Si work at Standard Telephone Laboratory (STL), England (1965-1969).

The work on a-Si using the “electrode- less glow discharge” (now referred to as PECVD¹) in silane gas (SiH₄) was pioneered by Sterling and his co-workers at STL starting in 1965 [2]. A seminal paper was published in 1969 by Chittick et al [3] and their findings summarized as follows: (i) *dark conductivity* (σ_d) *as a function of deposition temperature*, T_d : films fabricated in the range of T_d ~100°C to 500°C exhibited σ_d from $\sim 5 \cdot 10^{-10} \text{ (}\Omega\text{-cm)}^{-1}$ to $10^{-4} \text{ (}\Omega\text{-cm)}^{-1}$ while the conductivity activation energies in these samples ranged from 0.83eV to 0.2eV respectively. They recognized that the films were “ micro-crystalline or amorphous in form....”. (ii) *Photoconductivity*- σ_p : they noted that while σ_p was negligible for room temperature deposited samples, there was a pronounced change in σ_p of about 10^3 with 100ft candle illumination (corresponding to $\sim 100 \text{ mW-cm}^{-2}$) for samples deposited at 300°C. They also noted a smeared absorption edge between the wavelengths of 1.5 μm to 0.5 μm and reasoned that this was due to the then new idea of localized states at the band edges. (iii) *Aging or “instability”*: this was most noticeable for samples produced with T_d in the range of 200C -500C, i.e. the resistivity increased by about a factor of 50 after a few days- the first report of inherent degradation in a-Si ! (iv) *Doping*: n type doping was achieved by adding phosphine (0.4-4%) to the silane gas resulting in films with $\sigma_d \sim 10^{-4} \text{ (}\Omega\text{-cm)}^{-1}$, representing a change of $\sim 10^6$ from its intrinsic (undoped) value.

The “electrode- less glow discharge” deposition technique employed by the STL group was inductively coupled and the system would now be considered to be rudimentary at best, as it only used a rotary vane pump for pump down and processing, the quartz tube- in which decomposition was achieved- was held in place via simple O ring seals and the gas delivery as well the pumping used simple fittings. These aspects would have led to a high pre-deposition pressure, large leak rates resulting in contamination (e.g. O, C, N, pump oil etc.) of the samples. Given the system limitations, the data obtained on a-Si and the effects noted were quite remarkable. The work at STL on a-Si was terminated at the end of 1969.

2.0 Work at the University of Dundee from 1970 onwards. Doping and the field effect technique.

Professor Spear’s group in 1970 at Carnegie Laboratory of Physics, The University of Dundee, Scotland, UK consisted of Dr. Peter LeComber and research students with the laboratory situated in a converted jute shed. Spear had previously developed [4] a drift mobility (μ_d) technique which was particularly suitable for materials with high resistivity and low carrier mobility. (It should be noted that this μ_d technique is applicable in materials in which the dielectric relaxation time is very much longer than the transit time. This enables μ_d to be measured on both types of carriers in a semiconductor by merely reversing the field across the sample. The situation differs fundamentally from the drift of minority carriers in an extrinsic semiconductor where the condition of local space charge neutrality during carrier transit time forms a stringent requirement.) The primary focus at Dundee up to 1970 was the use of μ_d technique to study the transport of excess carriers in diatomic molecular solids and liquids, such as N₂, O₂ and CO. In 1969, Professor Spear became aware of the pioneering work of Professor’s Mott, Davis, Shaw, Cohen and Anderson [5] who introduced new concepts of electronic models of disordered materials, specifically band edges and band tails which

¹ PECVD: plasma enhanced chemical vapor deposition

should be inherent due to the structural disorder. While the majority of the work at that time on disordered semiconductors was on complex chalcogenide material systems, such SiTeAsGe, Spear reasoned that it would be more instructive to study a simpler material system, namely Si, such that through comparison with its crystalline counterpart, meaningful conclusions could be made. Hence in 1970, work started in Dundee on evaporated and sputtered Si and it quickly became apparent that as σ_d was quite high ($\sim 10^{-3} (\Omega\text{-cm})^{-1}$) and since the transport properties (hopping conductivity) were almost entirely determined by structural defects, it would then obscure any of the phenomena associated with inherent disorder (band tails). However, with the emergence of another type of a-Si (PECVD variety), as reported by the STL group, it represented more of a promising situation to study the band tails. In early 1970, LeComber and Spear [6] used their μ_d technique and employed a pulsed beam (10ns pulse width, 6 keV energy) of electrons to generate excess carriers in a 1 μm thick a-Si sample prepared using the inductively coupled PECVD technique. Transit of carriers (electrons but not holes) was observed in a pulsed applied field and synchronized to the excitation. μ_d and σ_d as function of temperature (T) for samples produced at $T_d \sim 200^\circ\text{C}$ revealed “kinks” as shown in Fig. 1; a model was proposed (which distinguished the PECVD material from the evaporated variety) with three transport paths, (a) for $T > 250\text{K}$, the conduction was attributed to transport above the conduction band edge, E_c , (b) for $T < 250\text{K}$, conduction occurred within the band tail states and (c) at a lower temperature still, hopping within the defect states deep in the μ -gap dominated [3]. The room temperature mobility, μ_d (for electrons), was measured to be $10^{-1}(\text{cm}^2 \text{ s}^{-1} \text{ V}^{-1})$ and the trap controlled mobility could be represented by $\mu_d = \mu_o \alpha \exp[-(E_c - E_t)/kT]$, where μ_o is the mobility near the bottom of E_c , and α depends on the form of the distribution of states below E_c controlling the drift mobility. They deduced that the conductivity mobility, μ_o was about $10 (\text{cm}^2 \text{ s}^{-1} \text{ V}^{-1})$ which supported the models of Mott and Cohen [3] who predicted a mobility of similar magnitude for transport of carriers through delocalized states where the mean path was on the order of the interatomic distance.

P to n transition in PECVD a-Si, photoconductivity and field effect.

I joined the Dundee group in August 1970 as a PhD student, with the specific task to use the above techniques ($\mu_d(T)$, $\sigma_d(T)$) to systematically study a-Si prepared by the inductively coupled PECVD and other techniques (evaporated and sputtered). Within a year or so, we had shown what appeared to be consistent picture for a-Si for samples prepared by the different techniques; for $T_d < 380K$ (PECVD) and for evaporated and sputtered Si samples, σ_d was dominated by hopping transport, as the pre-exponent, σ_0 , ($\sigma_d = \sigma_0 \exp(-E/kT)$) exhibited $10^{-4} (\Omega\text{-cm})^{-1}$ (corresponding to hopping conduction) while for samples produced for $T_d > 400K$ (PECVD) there was an abrupt change with $\sigma_0 > 100 (\Omega\text{-cm})^{-1}$ (corresponding to extended state conduction). μ_d measurements on the PECVD samples (irrespective of T_d) revealed that its value did not change substantially from $\sim 10^{-1} (\text{cm}^2 \text{s}^{-1} \text{V}^{-1})$ and that its activation energy (determined from $\mu_d(T)$) remained constant at $\sim 0.2\text{eV}$, corresponding to trapping occurring at the edge of the band tails states. We further reasoned that for $T_d < 380K$ (PECVD) and for evaporated a-Si, the samples were primarily p type (hopping) and for $T_d > 380K$ (PECVD), the samples exhibited n-type extended conduction [7]. In late 1971, it also become apparent that the PECVD a-Si presented an interesting material to study further, as the higher T_d produced samples exhibited very pronounced photoconductive behavior; at this point Dr. Ron Loveland switched over to the a-Si side of activities with the specific purpose to study the absorption coefficient and photoconductivity. Loveland et al reported [8] that for $T_d \sim 550K$ (PECVD), $X = i_p / eN_0(1-R) \sim 5 \cdot 10^{-1} (\text{A-s/C-cm}^2)$ where i_p is the photocurrent, N_0 is the number of photons and R is the reflection at the air/semiconductor interface. (I now estimate X leads to σ_p of about $10^{-4} (\Omega\text{-cm})^{-1}$ for an electric field of $3\text{-}10\text{KV/cm}$ applied to a coplanar sample under an illumination intensity of 100mW cm^{-2}). However, samples produced at $T_d \sim 370K$ (PECVD) and for evaporated Si, σ_p was orders of magnitude lower at $\sim 10^{-9} (\Omega\text{-cm})^{-1}$. We reasoned that the p to n transition with T_d (PECVD samples), coupled with high $\sigma_p \sim 10^{-4} (\Omega\text{-cm})^{-1}$ was due to the defect density being reduced as T_d was increased. Hence, the determination of the density and distribution

of states (DOS- $N(E)$) within the μ -gap would then be of crucial importance to understanding the results and tying it into band tail states picture. Spear and LeComber [9] undertook the field effect (FE) studies which had been previously employed to study surface states on crystalline semiconductor. (FE relies on applying a voltage across an insulator to induce charge into the semiconductor creating either an accumulation layer or a depletion layer and thus altering the current, I_{sd} , between the source and drain contacts. The extent of the band bending is inextricably linked to the DOS around the Fermi level, E_f .) As shown in Fig. 2, they reported that $N(E_f)$ was extremely low and approximately $10^{15} \text{ cm}^{-3} \text{ eV}^{-1}$ for samples produced at $T_d \sim 630\text{K}$, while for $T_d \sim 400\text{K}$, the DOS increased significantly, $N(E_f) \sim 10^{17} \text{ cm}^{-3} \text{ eV}^{-1}$; the evaporated samples exhibited large DOS, $N(E_f) > 10^{19} \text{ cm}^{-3} \text{ eV}^{-1}$. In 1972 [10], we presented a cohesive model based on drift mobility, conductivity, photoconductivity and the field effect data which provided further experimental proof provided earlier by Spear and LeComber [6] of Mott's ideas .i.e. there were two distinct groups of localized states, (a) a distribution adjoined the extended states from E_a to E_c (critical energy corresponding to the conduction band) and between E_v (critical energy corresponding to the valence band) and E_b which originate from the lack of long range order. While the extent of $E_c - E_v$ was thought to be 1.55eV, the extent of band tails, $E_c - E_a$, was surmised to be about 0.2eV, (b) the localized states in the central region between E_a and E_b which are likely associated, in an elemental semiconductor with reasonably defined structural defects. These conclusions were derived on the basis that (i) as the activation energy of about 0.2eV from $\mu_d(T)$ data, did not change with T_d implying that E_a remained constant, while (ii) the conductivity as function of T_d changed drastically suggesting the movement of E_f is determined by the structural defects. This seemed to be corroborated from the FE results for samples deposited at different T_d , in which it was shown that as T_s is lowered the DOS increases as shown in Fig. 2.

However the FE results discussed above involved the use of soda lime glass as the dielectric and it was soon discovered that the amount of charge being induced into the a-Si was about a factor of 6

larger (due to ionic contribution from the soda lime glass) than accounted for in the calculations which in turn would have implied that the $N(E_f)$ was actually larger than indicated. Further, as Spear and LeComber had noted a large hysteresis effect (i.e. I_{sd} showed different values when the gate voltage, V_g , was increased and then reduced to zero) then there were obvious problems with the FE technique as used. Although I had essentially completed the work for my Ph.D studies, I stayed on for another year (with some reluctance, I might add) to work on FE technique to try and correct the problems with the technique and analysis. In the first instance, we needed to substitute soda lime glass with quartz in which ionic contribution would be absent. As the quartz was quite thick (175 μm) and in order to induce enough charge into the a-Si (to establish an accumulation layer), very high voltage ($> 15\text{kV}$) was needed, as shown in Fig. 3a; as was inevitable, the use of such high voltage on occasion led to arcing (even in vacuum) resulting in damage to electronic equipment and putting a severe dent in the meager budget of the group. Hence it became incumbent upon the author to evaluate and develop the use of other dielectrics, specifically thin SiN_x using the PECVD technique and prepared from SiH_4 and N_2 gasses. This was developed during the early part of 1973 [11,12] and using a structure of the type, gate-metal/ SiN_x /a-Si and aluminum source and drain contacts, led to I_{on}/I_{off} ratio $\sim 10^4$ for a gate voltage of $< 40\text{V}$ using SiN_x thickness in the range of 1-3 μm [5], as shown in Fig. 3b. From the FE data using quartz and SiN_x data, $N(E_f)$ was established to be higher at approximately $10^{17} \text{ cm}^{-3} \text{ eV}^{-1}$, as shown in Fig.3(c). Use of the combination a-Si/ SiN_x , is the basis of a TFT (thin film transistor) which are used to switch pixels in AMLCD.

Substitutional Doping and p-n Junction.

On Sept 5th 1972, an internal memo (which I still have) was created and distributed to the group by Peter LeComber which detailed the future strategy and what needed to be done with a-Si based on our understanding at the time. Some of the main features of the memo were, (a) establish doping (n and p type), (b) produce p-n junctions, (c) produce p-i-n junctions, (d) evaluate the use of capacitive

coupling of plasma etc. To dope using B_2H_6 (diborane) and PH_3 gasses, was a major concern to all of us because no one at Dundee had experience with the use of such highly toxic and poisonous materials. In the memo, it was stated that as B_2H_6 is “extremely poisonous—leave at present”. It was not until 1975 that Spear and LeComber [1] published the landmark paper and results are shown in Fig. 4; by adding PH_3 to the SiH_4 gas resulted in films whose σ_d changed from its intrinsic value of $\sim 10^{-9} (\Omega\text{-cm})^{-1}$ to $\sim 10^{-2} (\Omega\text{-cm})^{-1}$ with N_{PH_3}/N_{SiH_4} (in the gas phase) of $\sim 10^{-3}$. Corresponding activation energy measurements revealed that the E_f continuously shifted to within 0.15eV of the conduction band edge, E_c . They concluded “...that an appreciable density of the pentavalent impurities could be incorporated substitutionally into the random network where they produce donor states within 0.1eV of the extended state electrons”. For the boron doped samples, with $N_{B_2H_6}/N_{SiH_4}$ up to $\sim 2 \cdot 10^{-5}$, the predominant conduction path remained in the extended states. As E_f moved past the states at the minimum in DOS at E_c-E_f , the centre of the μ -gap, the samples exhibited suddenly a p-type behavior and σ_d rose by several orders of magnitude to about $\sim 10^{-2} (\Omega\text{-cm})^{-1}$ for $N_{B_2H_6}/N_{SiH_4} \sim 10^{-2}$. They concluded “...that contrary to thought at the time that it was indeed possible to incorporate substitutionally pentavalent and trivalent impurities into the tetrahedral amorphous semiconductors, such as Si”. Spear and Le Comber adhered to their strategy formulated in 1972 and very quickly followed their landmark paper with the report of the first p-n junction in early 1976, clearly showing rectification [13]. Several months later, Carlson et al [14] inserted the highly photoconductive intrinsic layer (already established by Loveland et al [8]) into a p-i-n junction configuration and produced a solar cell with a 2.4% conversion efficiency. In 1981, Le Comber [15] integrated the FE transistors (or TFT's) into a complete functioning 7 X 5 array; commercialization took root in the 1980's, especially in Japan which has this year culminated into a 65 inch AMLCD television reported by Sharp Corporation.

Space prevents me from recognizing the work of David Adler, Bob Street, J. Stuke, H. Fritzsche, G. Lucovsky, Marc Brodsky and Bill Paul who played a crucial role in elucidating defects and the role

of H in the a-Si fabricated from the PECVD technique using SiH_4 gas. I should like to also mention major contributions from a commercialization point of view, specifically (a) Knights et al [16], who showed that doped and undoped a-Si could be fabricated using a capacitively coupled system and the film properties (undoped and doped) were equivalent to the inductively coupled PECVD systems and (b) Kuwano et al reported the use of separated reaction chambers [17] which eliminated the cross contamination between intrinsic and doped layers, necessary for high performance devices. These two aspects have led to reproducible large area depositions of electronic devices such as TFT's and solar cells etc. These types of systems are now used routinely in industry and referred to as multi-chamber or cluster tools.

Future of Amorphous Silicon.

Although a-Si appears to be a mature technology, it is an incredible testament to the versatility of this basic material that new applications continue to emerge such as memory devices, thermal imaging using filters, spatial light modulators, solar to hydrogen conversion, etc. Where can the a-Si technology make a large impact in the future? A major application on the horizon could be flexible displays: the use of low temperature ($\sim 150^\circ\text{C}$) during processing and the ability to manipulate the growth etc. allows the use of a TFT on low cost flexible plastic materials, such as PEN and PET. Professor Arokia Nathan's group have already reported a TFT deposited on plastic substrate at 150°C with $I_{\text{on}}/I_{\text{off}} > 10^7$ [18]. They have also reported on nano-crystalline Si NMOS and PMOS structures with field effect mobility of electrons and holes of ~ 200 and $\sim 35 (\text{cm}^2 \text{ s}^{-1} \text{ V}^{-1})$ respectively [19]. The flexible display application represents another multi-billion dollar application, although competition may come from organic semiconductors. Given the history of a-Si and the amount of investments made, a-Si TFT's for this application may prove to be the winner.

Another major application area is in the field of renewable energy, i.e solar cells or photovoltaics (PV). Renewable energy benefits are not only obvious but represent a trillion dollar business potential and could dwarf anything in the electronics area. The market PV in 2004 was ~1300 Megawatts at < \$2.50/Watt and it is expanding at the rate of 20-30% per year. Crystalline Si/multi-crystalline Si solar cells presently dominate the market with thin film Si share at <10%. At the moment, a-Si is represented primarily in niche markets, (calculators, battery chargers, applications in building integrated PV etc.). The efficiency of a-Si based commercial modules is < 7% (with cost > crystalline Si solar cell) and production levels somewhat insignificant in relation to its crystalline counterpart. There are numerous issues associated with this state of affairs such as, low deposition rate, low gas utilization, use of prohibitively expensive gasses such as GeH₄, use of complex current constraining two terminal tandem junctions (instead of perhaps 4-terminal devices [20]) which do not entirely mitigate against the inherent degradation of a-Si. The challenge for thin film silicon to truly realize its potential is to break out of this straight jacket otherwise it may remain marginalized in the rapidly expanding PV market.

Where is the original Dundee team now?

In 1977, I moved to Energy Conversion Devices, Michigan, USA (which had a total employee count of 20) to initiate the work on a-Si and produced a-SiFH alloy [21] which generated much interest, controversy and funding for ECD (~\$40million - not inflation adjusted) due primarily to Stanford R. Ovshinsky's tireless promotional efforts which launched that company into the a-Si arena. I left ECD in early 1982 and am the owner of MVSystems Inc. and a Research Professor at The Colorado School of Mines. Dr. Ron Loveland left Dundee in 1974 for ERA Technology to work on electrostatics research. After various stints with the Civil Service in the UK, he is currently the Chief Technology Officer in Welsh Assembly Government with special responsibilities in clean (low carbon) energy field-including developing PV in Wales. From 1975

until his retirement in the 1991, Spear (and LeComber) continued to provide major contributions from fundamental understanding of a-Si to developing high current diodes, memory devices etc. Walter Spear still lives in Dundee and is becoming fluent in the Italian language; we remain in touch with occasional letters criss-crossing the Atlantic. Peter Le Comber died at a very young age of 51 in 1993 of heart attack in Switzerland while commemorating his 30th wedding anniversary. His wife Joy still lives in Dundee.

Finally we owe a debt of gratitude to Professor Walter Spear and Dr. Peter Le Comber as they laid the foundation which was not only long lasting but provided the basis for a multi billion dollar industry with future growth assured.

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Figure captions.

1. Temperature dependence of (a) electron drift mobility, μ_d . (b) dark conductivity, σ_d in the “glow discharge” films of a-Si deposited at 500K. The inset in the top figure shows a model of the electronic state distribution in an amorphous solid [6].
2. The density of localized states, $N(E)$, plotted against the energy E_c-E for a number of a-Si samples produced using the “glow discharge” (or PECVD) technique in SiH_4 gas. The temperature T_d refers to the deposition temperature. ESi refers to an evaporated Si sample. L and S denote linear and square law interpolations respectively [9].
3. The dependence of the sample source-drain current, I_d on the gate potential V_g for a number of “glow discharge” (or PECVD) samples deposited at about 250C using (a) SiN_x dielectric and (b) quartz dielectric [11,12]. (c) Deduced density of states, $N(E)$, for “glow discharge” (or PECVD) specimens deposited at the given temperature, T_d , using the SiN_x or the quartz dielectric. The position of the Fermi level, E_f , is indicated by the arrow. Full lines were calculated from the $I_d(V_g)$ curves and the dotted parts were based on an extrapolation procedure. [11,12].
4. The effect of phosphorous and boron doping on the conductivity and the position of the Fermi level in a-Si specimens. On the right these quantities are plotted against the phosphine concentration in the gas mixture used for the specimen preparation and on the left against the borane concentration. The center refers to undoped specimens [1].

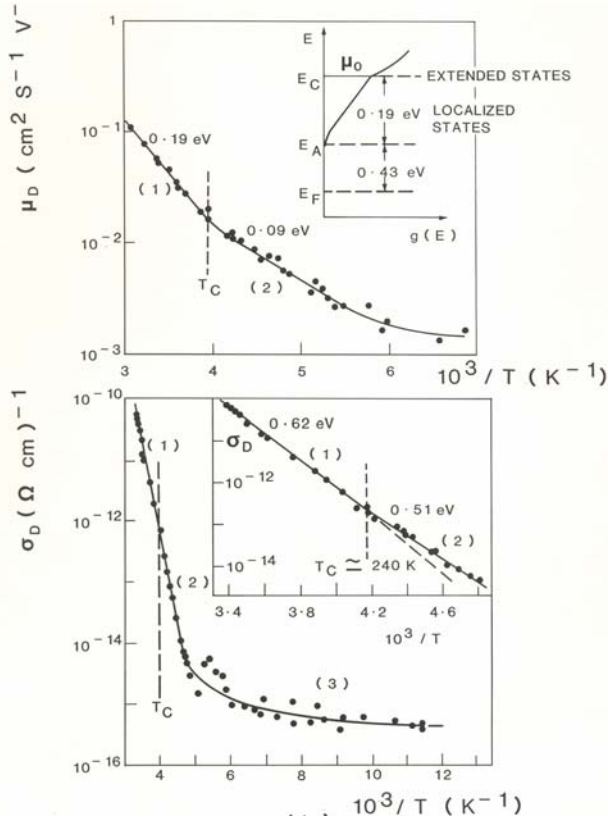


Fig. 1

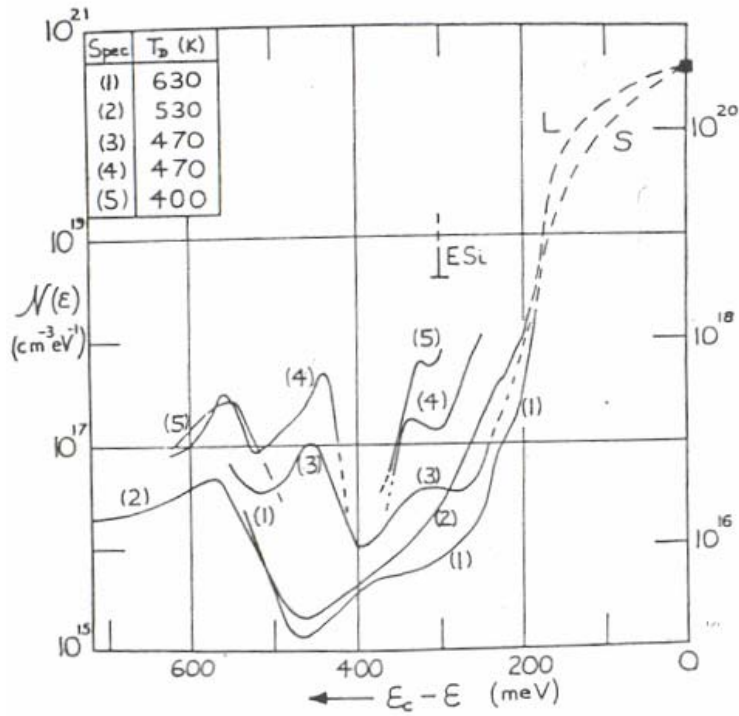


Fig 2

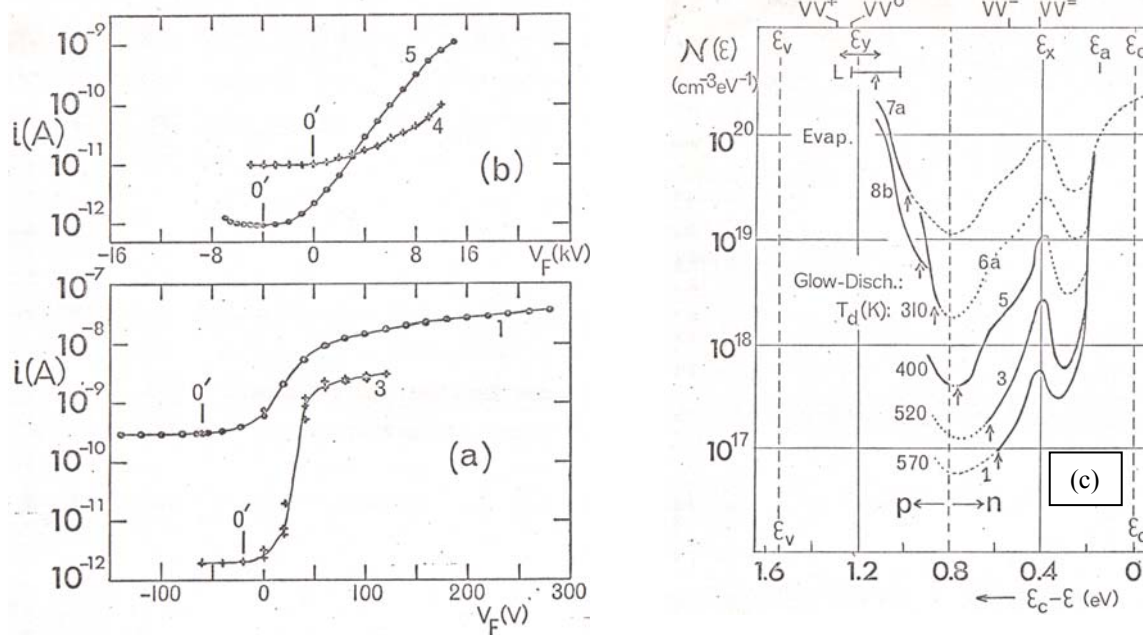


Fig. 3

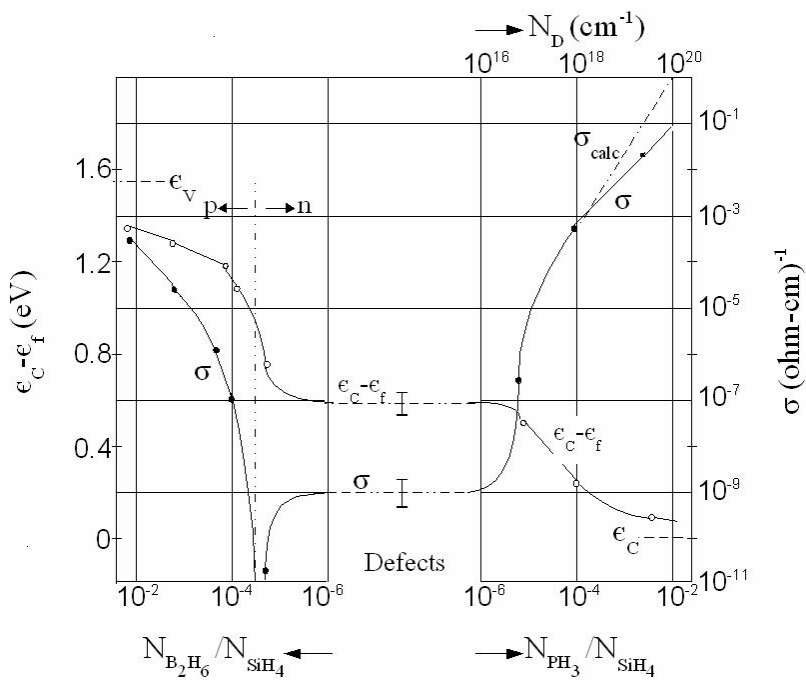


Fig. 4